JC17 Rec'd PCT/PTO 02 MAY 2001

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35.U.S.C. 371	EXPRESS MAIL LABEL No EF321689252US	DATE May 2, 2001
	ATTORNEY'S DOCKET NO. A34258-PCT-USA-A - 072035.0122	
	US APPLICATION NO 09/830897	

INTERNATIONAL APPLICATION NO	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/GB99/03619	2 November 1999	2 November 1998

TITLE OF INVENTION
ELECTROLUMINESCENT MATERIALS

APPLICANT(S) FOR DO/EO/US
Poopathy Kathirgamanathan

Applicant herewith submits to the United States Designated /Elected Office (DO/EO/US) the following items and other information:

. [X] This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
. [ ] This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
. [X] This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination ntil the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(I).
. [X] A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date
. [X] A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  a. [ ] is transmitted herewith (required only if not transmitted by the International Bureau).
  b. [X] has been transmitted by the International Bureau.
  c. [ ] is not required, as the application was filed in the United States Receiving Office (RO/US).
[ ] A translation of the International Application into English (35 U.S.C. 371(c)(2)).
[X] A copy of the International Search Report (PCT/ISA/210)
  a. [ ] are transmitted herewith (required only if not transmitted by the International Bureau).
  b. [X] have been transmitted by the International Bureau
  c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
  d. [ ] have not been made and will not be made.
[ ] A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
[X] An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
[ ] A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

ms 11. to 16. below concern other document(s) or information included:

[X] A copy of the International Preliminary Examination Report (PCT/IPEA/409)
[X] An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
[X] A FIRST preliminary amendment.
[ ] A SECOND or SUBSEQUENT preliminary amendment.
[ ] A substitute specification.
[ ] A change of power of attorney and/or address letter.
[X] Other items or information:
  a. [ ] a copy of the International Search Report (PCT/ISA/210)
  b. [ ] a copy of the International Preliminary Examination Report (PCT/IPEA/409)
  c. [X] PCT application No. PCT/GB99/03619 was published in English under publication number WO 00/26323 on May 11, 2000. The present invention relates in general to the field of microwave devices and artificial dielectrics

INTERNATIONAL APPLICATION NO. PCT/GB99/03619	INTERNATIONAL FILING DATE 2 November 1999	PRIORITY DATE CLAIMED 2 November 1998

09/830897

7. [X] The following fees are submitted:

Basic National Fee (37 CFR 1.492(a)(1)-(5):

Neither international preliminary examination fee (37 CFR 1.482)

Nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO (1.492(a)(3)) $1,000.00

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO (1.492(a)(5) $860.00

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO(1.492(a)(2)) $710.00

International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) (1.492(a)(1)) $690.00

International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) $ 100.00

CALCULATIONS PTO USE ONLY

ENTER APPROPRIATE BASIC FEE AMOUNT				=	$860.00
charge of $130.00 for furnishing the oath or declaration later than [ ] 20 [ ] 30 ths from the earliest claimed priority date (37 C.F.R. 1.492(e)).					$130.00
Claims	**Number Filed**		**Number Extra**	**Rate**	$
al Claims	17	-20=		X $ 18.00	$
ependent Claims	1	-3=		X $ 80.00	$
ltiple dependent claim(s) (if applicable)				+ $270.00	$
TOTAL OF ABOVE CALCULATIONS				=	$990.00
duction by ½ for filing by small entity, if applicable					$
SUBTOTAL				=	$
cessing fee of $130.00 for furnishing the English translation later than [ ] 20 [ ] 30 nths from the earliest claimed priority date (37 CFR 1.492(f)). +					$
TOTAL NATIONAL FEE				=	$
for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be ompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). $40.00 per property +					$ 40.00
TOTAL FEES ENCLOSED				=	$1030.00
					Amt. refunded $
					charged $

[X] A check in the amount of $1030.00 to cover the above fees is enclosed.

[ ] Please charge our Deposit Account No. 02-4377 in amount of $___ to cover the above fees. A copy of this sheet is enclosed.

[X] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 02-4377. A copy of this sheet is enclosed.

TE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or) must be filed and granted to restore the application to pending status.

ND ALL CORRESPONDENCE TO:

KER BOTTS L.L.P.
Rockefeller Plaza
w York, New York 10112-4498

Marta E Delsignore
Signature

May 2, 2001
Date

32,689
Registration No.

02:322479.1

A34258-PCT-USA - 072035.

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Poopathy Kathirgamanathan

Serial No. : To be assigned

Filed : Herewith

For : NOVEL ELECTROLUMINESCENT MATERIALS

PRELIMINARY AMENDMENT

I hereby certify that this paper is being deposited with the United States Postal Service as Express Mail in an envelope addressed to: Assistant Commissioner for Patents, Washington, D.C. 20231

May 2, 2001
Date of Deposit

Marta E. Delsignore
Attorney Name

Marta E. Delsignore
Signature

32,689
PTO Registration No.

May 2, 2001
Date of Signature

EXPRESS MAIL NO.: EF321689252US

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Preliminary to examination, please amend the above-identified patent application as follows.

IN THE SPECIFICATION:

Please replace the paragraph beginning at page 1, line 1, with the following rewritten paragraphs:

--This application is a national stage application of PCT/GB99/03619 which was published in English under publication number WO 00/26323.

The present invention relates to electroluminescent materials and to devices incorporating them.--

At page 1, after line 5, please insert the following:

--BACKGROUND OF THE INVENTION--

At Page 2 after line 6, please insert the following:

--SUMMARY OF THE INVENTION--

At page 2, after line 20, please insert the following:

--BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a chromacity diagram;

Fig. 2 is an electroluminescent spectrum for a device prepared in accordance with Example 1;

Fig. 3 is an electroluminescent spectrum for a device prepared in accordance with Example 2;

Fig. 4 is an electroluminescent spectrum for a device prepared in accordance with Example 3;

Fig. 5 is an electroluminescent spectrum for a device prepared in accordance with Example 4; and

Fig. 6 is an electroluminescent spectrum for a device prepared in accordance with Example 5.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS--

Please replace in the paragraph beginning at page 1, line 1, with the following rewritten paragraph:

--I CLAIM:--

IN THE CLAIMS:

Please amend the claims as follows:

1. (Amended) An electroluminescent compound which comprises an organic complex of a metal and an organic ligand which emits light in the blue or purplish blue spectrum when an electric current is passed through it

wherein the metal is selected from the group consisting of thorium (IV), yttrium (III), gadolinium (III), europium (II), terbium (IV), cerium (IV), cerium (III) and mixtures thereof and the ligand is selected from the group consisting of

and

where R' maybe the same or different at different parts of the molecule and each of R" and R' is a substituted or unsubstituted aromatic or heterocyclic ring structure or a hydrocarbyl or a fluorocarbon or R" is fluorine or hydrogen or R" is copolymerised with a monomer or R' is t-butyl and R" is hydrogen.

Please cancel claims 2-5.

6. (Amended) An electroluminescent compound according to claim 1 having the formula $Eu(II)(TMHD)_2$.

7. (Amended) A composition which comprises an inert polymer and from 5% to 95% by weight of an electroluminescent compound as claimed in claim 1.

8. (Amended) An electroluminescent device which comprises (i) a transparent substrate (ii) an electroluminescent layer comprising an electroluminescent compound as claimed in claim 1 deposited on the substrate and (iii) a cathode.

Please cancel claim 10.

11. (Amended) An electroluminescent device as claimed in claim 8 in which there is a hole transporting layer deposited on the transparent substrate and the electoluminescent material is deposited on the hole transporting layer.

12. (Amended) An electroluminescent device as claimed in claim 8 in which there is a hole transporting material mixed with the electroluminescent material in a ratio of 5 to 95% of the electroluminescent material to 95 to 5% of the hole transporting compound.

13. (Amended) An electroluminescent device as claimed in claim 12 in which the hole transporting material is an aromatic amine complex.

14. (Amended) An electroluminescent device as claimed in claim 13 in which the hole transporting material comprises at least one selected from the group consisting of poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1' -biphenyl -4,4' diamine (TPD) and polyaniline.

Please cancel claim 15.

16. (Amended) An electroluminescent device as claimed in claim 8 in which there is a layer of an electron injecting material between the cathode and the electroluminscent material layer.

17. (Amended) An electroluminscent device as claim in claim 8 wherein the electroluminescent layer includes an electron injecting material.

18. (Amended) An electroluminescent device as claimed in claim 16 wherein the electron injecting material is a metal complex or oxadiazole or an oxadiazole derivative.

20. (Amended) An electroluminscent device as claimed in claim 8 wherein the electroluminescent layer includes a dye.

Please cancel claim 22.

23. (Amended) An electroluminescent device as claimed in claim 8 in which the anode includes one selected from the group consisting of aluminum, magnesium, lithium, calcium and magnesium silver alloy.

24. (Amended) An electroluminescent device as claimed in claim 8 comprising a plurality of electroluminescent layers.

25. (Amended) An electroluminescent device as claimed in claim 8 wherein the electroluminescent layer comprising at least two electroluminescent compounds.

Please add the following new claims.

26. (New) An electroluminescent device as claimed in claim 11 wherein the hole transporting layer comprises an aromatic amine complex.

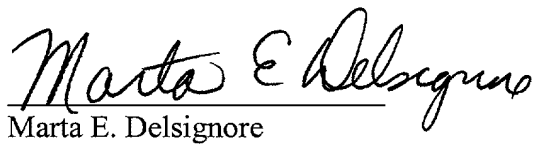
27. (New) An electroluminescent device as in claim 11 wherein the hole transporting comprises at least one selected from the group consisting of poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1' -biphenyl -4,4' diamine (TPD) and polyaniline.

REMARKS

By the foregoing amendment, the specification and claims have been amended to conform more closely to U.S. patent practice.

Attached hereto is a marked-up version of the changes made to the specification and claims by the current amendment. The attached page is captioned "**Version with markings to show changes made.**"

Respectfully submitted,



Marta E. Delsignore
Patent Office Reg. No. 32,689

Attorney for Applicants

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New York, NY 10112
(212) 408-2632

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION:

Please replace the paragraph beginning at page 1, line 1, with the following rewritten paragraphs:

--This application is a national stage application of PCT/GB99/03619 which was published in English under publication number WO 00/26323.

The present invention relates to electroluminescent materials and to devices incorporating them.--

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Fig. 1 is a chromacity diagram;

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Fig. 3 is an electroluminescent spectrum for a device prepared in accordance with Example 2;

Fig. 4 is an electroluminescent spectrum for a device prepared in accordance with Example 3;

Fig. 5 is an electroluminescent spectrum for a device prepared in accordance with Example 4; and

Fig. 6 is an electroluminescent spectrum for a device prepared in accordance with Example 5.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS--

Please replace in the paragraph beginning at page 1, line 1, with the following rewritten paragraph:

--I CLAIM:--

IN THE CLAIMS:

Please amend the claims as follows:

1. (Amended) [A photoluminescent] An electroluminescent compound which comprises an organic complex of a [transition] metal[, a lanthanide or an actinide] and an organic ligand which [photoluminescent compound] emits light in the blue or purplish blue spectrum when an electric current is passed through it

wherein the metal is selected from the group consisting of thorium (IV), yttrium (III), gadolinium (III), europium (II), terbium (IV), cerium (IV), cerium (III) and mixtures thereof and the ligand is selected from the group consisting of

and

where R' maybe the same or different at different parts of the molecule and each of R" and R' is a substituted or unsubstituted aromatic or heterocyclic ring structure or a hydrocarbyl or a fluorocarbon or R" is fluorine or hydrogen or R" is copolymerised with a monomer or R' is t-butyl and R" is hydrogen.

Please cancel claims 2-5.

6. (Amended) An electroluminescent compound according to claim 1 having the formula $Eu(II)(TMHD)_2$.

7. (Amended) A composition which comprises an inert polymer and from 5% to 95% by weight of an electroluminescent compound as claimed in [any one of the preceding claims] claim 1.

8. (Amended) An electroluminescent device which comprises (i) a transparent substrate [on which is deposited] (ii) an electroluminescent layer comprising an electroluminescent compound as claimed in [any of the preceding claims] claim 1 deposited on the substrate and (iii) a cathode.

Please cancel claim 10.

11. (Amended) An electroluminescent device as claimed in [any one of claims 8 to 10] claim 8 in which there is a hole transporting layer deposited on the transparent substrate and the electoluminescent material is deposited on the hole transporting layer.

12. (Amended) An electroluminescent device as claimed in claim [11] 8 in which there is a hole transporting material mixed with the electroluminescent material in a ratio of 5 to 95% of the electroluminescent material to 95 to 5% of the hole transporting compound.

13. (Amended) An electroluminescent device as claimed in claim 12 in which the hole transporting [layer] material is an aromatic amine complex.

14. (Amended) An electroluminescent device as claimed in claim 13 in which the hole transporting [layer is] material comprises at least one selected from the group consisting of poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1' -biphenyl -4,4' diamine (TPD) [or] and polyaniline.

Please cancel claim 15.

16. (Amended) An electroluminescent device as claimed in [any one of claims 8 to 15] claim 8 in which there is a layer of an electron injecting material between the cathode and the electroluminscent material layer.

17. (Amended) An electroluminscent device as claim in [any one of claims 8 to 16 in which an electron injecting material is mixed with] claim 8 wherein the electroluminescent layer includes an electron injecting material [and co-deposited it].

18. (Amended) An electroluminescent device as claimed in claim 16 [or 17 in which] wherein the electron injecting material is a metal complex or oxadiazole or an oxadiazole derivative.

20. (Amended) An electroluminscent device as claimed in [any one of claims 8 to 19 in which there is a dye incorporated in] claim 8 wherein the electroluminescent layer includes a dye.

Please cancel claim 22.

23. (Amended) An electroluminescent device as claimed in claim [22] 8 in which the anode includes one selected from the group consisting of [is a] aluminum, magnesium, lithium, calcium [or] and magnesium silver alloy.

24. (Amended) An electroluminescent device as claimed in [any one of the preceding claims in which there are] claim 8 comprising a plurality of electroluminescent layers [of electroluminescent material].

25. (Amended) An electroluminescent device as claimed in [any one of the preceding claims in which] claim 8 wherein the electroluminescent layer [of electroluminescent material is formed of] comprising at least two [or more different] electroluminescent compounds.

ELECTROLUMINESCENT MATERIALS

The present invention relates to electroluminescent materials and to devices incorporating them.

Materials which emit light when an electric current is passed through them are well known and used in a wide range of display applications. Liquid crystal devices and devices which are based on inorganic semiconductor systems are widely used. however these suffer from the disadvantages of high energy consumption, high cost of manufacture, low quantum- efficiency and the inability to make flat panel displays. reflectance problems, i.e. low visibility in bright conditions and a narrow viewing angle e.g. +/- 45°.

Organic polymers have been proposed as useful in electroluminescent devices, but it is not possible to obtain pure colours, they are expensive to make and have a relatively low efficiency.

Another compound which has been proposed is aluminium quinolate, but this requires dopants to be used to obtain a range of colours and has a relatively low efficiency.

In an article in Chemistry letters pp 657-660, I990 Kido et al disclosed that a terbium (III) acetyl acetonate complex was green electroluminescent and in an article in Applied Physics letters 65 (17) 24 October I994 Kido et al disclosed that a europium (III) triphenylene diamine complexes was red electroluminescent but these were unstable in atmospheric conditions and difficult to produce as films.

The complexes disclosed in these articles had a relatively low photoluminescent efficiency and were only able to produce green or red light and other colours could not be produced.

We have now discovered photoluminescent and electroluminescent compounds and materials which emit blue and purplish blue light.

According to the invention there is provided an photoluminescent compound which comprises an organic complex of a transition metal, lanthanide or an actinide and an organic ligand which photoluminescent compound emits light in the blue or purplish blue spectrum.

It has surprisingly been found that it is possible by choice of the metal and the organic ligand to form a complex which, when an electric current is applied across it will emit blue or purplish blue light.

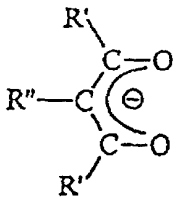
The invention also provides an electroluminescent compound which comprises an organic complex of a transition metal, a lanthanide or an actinide and an organic ligand which electroluminescent compound emits light in the blue or purplish blue spectrum when an electric current is passed through it.

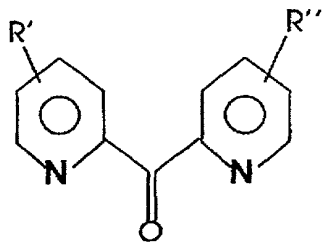
The colour of light is subjective and colours can be defined by co-ordinates on a two dimensional chart in which colours are areas on the chart and in the present invention the blue and purplish blue spectrum is defined as the area bounded by the co-ordinates in the colour chart CIE 1931 a copy of which is shown in Fig. 1. The complexes of the invention enable light within the co-ordinates (0, 0) (0, 0.3) (0.3, 0) to be produced.

Light in the blue region of the spectrum is difficult to produce and hitherto it has not been possible to produce blue light by means of electroluminescence.

The preferred metals are thorium (IV), yttrium (III), gadolinium (III), europium (II). terbium(IV), cerium(IV) and cerium (III). A mixture of metals can be used to form mixed chelates.

The preferred ligands are





or

where R′ is the same or different at different parts of the molecule and each R′′ and R′ is a substituted or unsubstituted aromatic or heterocyclic ring structure or a hydrocarbyl or a fluorocarbon or R′′ is fluorine or hydrogen or R′′ is copolymerised with a monomer e.g. or R′ is t-butyl and R′′ hydrogen.

Preferably each of R', R", and R' is an alkyl group preferably a -C(CH3) group,

Preferred complexes are TMHD (Tris(2,2,6,6-tetramethyl-3,5-heptanedionato). α′, α′′, α′′′ tripyridyl, bathophen (4,7-diphenyl-1,1-phenanthroline), crown ethers and cryptans.

Particularly preferred complexes are the thorium bathophen, yttrium tripyridyl and TMHD, and europium TMHD complexes.

A novel complex with strong photoluminescent and electroluminescent properties is $Eu(II)(TMHD)_2$ complex which is stable in air. It would have been expected that such a Eu(II) complex would have been unstable in the presence of oxygen and it is surprising that this complex is air stable.

The complexes of the present invention can be used to form electroluminescent devices.

The electroluminescent devices of the invention comprise a transparent substrate which is a conductive glass or plastic material which acts as the anode, preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass which is conductive or has a conductive layer can be used. Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the substrate. The electroluminescent material can be deposited on the substrate directly by evaporation from a solution of the material in an organic solvent. The solvent which is used will depend on the material for example alcohols such as ethanol. ketones such as acetone and methyl acetylacetonate, and chlorinated hydrocarbons such as dichloromethane are suitable in many cases.

Alternatively the material can be deposited by spin coating or by vacuum deposition from the solid state e.g. by sputtering or any other conventional method can be used.

In one embodiment of the invention there is a hole transporting layer deposited on the transparent substrate and the electroluminescent material is deposited on the hole transporting layer. The hole transporting layer serves to transport; holes and to block the electrons, thus preventing electrons from moving into the electrode without

recombining with holes. The recombination of carriers therefore mainly takes place in the emitter layer.

Hole transporting layers are used in polymer electroluminescent, devices and any of the known hole transporting materials in film form can be used.

The hole transporting layer can be made of a film of an aromatic amine complex such as poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1'-biphenyl -4,4'-diamine (TPD), polyaniline etc.

Optionally dyes such as fluorescent laser dyes, luminescent laser dyes etc. can be included to modify the colour spectrum of the emitted light.

Preferably the electroluminescent material is mixed with an inert polymeric material such as a polyolefin e.g. polyethylene, polypropylene etc. and preferably polystyrene. Preferred amounts of the electroluminescent material in the mixture is from 95% to 5% by weight of active material and more preferably 25 to 20% by weight.

The hole transporting compound can optionally be mixed with the electroluminescent material in a ratio of 5-95% of the electroluminescent material to 95 to 5% of the hole transporting compound. In another embodiment of the invention there is a layer of an electron injecting material between the cathode and the electroluminescent material layer, this electron injecting material is preferably a metal complex such as a metal quinolate e.g. an aluminium quinolate which will transport electrons when an electric current is passed through it. Alternatively the electron injecting material can be mixed with the electroluminescent material and co-deposited with it.

In a preferred structure there is a substrate formed of a transparent conductive material which is the anode on which is successively deposited a hole transportation

layer, the electroluminescent material layer and an electron injection layer which is connected to the anode. The anode can be any low work function metal e.g. aluminium, calcium, lithium, silver/magnesium alloys etc.,

The preparation of compounds of the invention are shown in the examples

Example 1

Mono(bathophenanthroline)thorium(IV)chloride. Thorium(IV)chloride (5 mmol. 1.87 g) was dissolved in ethanol/water mixture (2:1 v/v) (75 ml) at 50'C. Bathophenanthroline (5 mmol, 1.66 g) was dissolved in a mixture of ethanol/dichloromethane (2:1 v/v) (75 ml) and added portionwise to the solution of the thorium salt. The mixture was reduced on a hotplate at 100°C over one hour. The precipitate was filtered to give an off-white solid which was washed with diethylether (2 x 25 ml) and dried in vacuo to yield the product (1.9 g).

Example 2

Mono(α', α'', α'''tripypyridyl)yttrium(III) chloride. Yttrium(III) chloride (2 mmol. 0.61 g) was dissolved in ethanol (100 ml) and α', α'', α''' tripyridyl (2 mmol, 0.47 g) was added. The reaction mixture was warmed for 60 minutes at 50°C and the solvents removed. The residue was washed with diethylether (2x25 ml) and dried in vacuo to give the product (0.80g).

Example 3

Tris(2,2,6,6-tetramethyl-3,5-heptanedionato)yttrium(III) mono(α′,α′′,α′′′tripyridyl). The tris-chelate (1 mmol. 0.64 g) was dissolved in ethanol (100 ml) and α′, α′′, α′′′tripyridyl (1 mmol, 0.23g) was added. The reaction mixture was warmed for 60 minutes at 50°C and the solvents removed. The residue was washed with diethylether (2x25 ml) and dried in vacuo to give the product (0.50 g). Yield 57%.

Example 4

Mono(α′, α′′, α′′′tripyridyl)gadolinium(III) chloride. Gadolinium(III) chloride (0.37 g, 1 mmol) was dissolved in ethanol (150 ml) and α′, α′′, α′′′-tripyridyl (0.23 g, 1 mmol) was added. The reaction mixture was heated under reflux for 1 hour and the solvent removed in vacuo to give the gadolinium adduct (Yield 0.50 g).

Example 5

Bis(2,2,6,6-tetramethyl-3,5-heptanedionato)europium(II). The reaction was carried out under anhydrous conditions using dried glassware under dry nitrogen and using acetyl nitrile freshly distilled over phosphorus pentoxide. Europium(II) chloride (1.0 g, 5 mmol) was placed in a 250 ml three-neck round-bottom flask fitted with a condenser, two dropping funnels and nitrogen bubbler. Deoxygenated solution of the diketone (1.84 gms 10 mmol) in acetyl nitrile (50 ml) was placed in the first dropping funnel and deoxygenated acetyl nitrile (150 ml) was placed in the second dropping funnel. Both funnels were under nitrogen and fitted with nitrogen balloons. Acetyl nitrile was allowed to run into the flask and the mixture stirred at 50°C (oil bath) until dissolution. The diketone solution was then added to the flask and the reaction mixture refluxed for one hour and allowed to cool under nitrogen overnight. The dry diethylether and dried in vacuo at 60°C. (Yield 0.82 g). The filtrate was dried to give pale yellow tris(2,2,6,6-tetramethyl-3,5-heptaneionato)europium(II). (Yield 0.90 g).

Electroluminescent devices were fabricated and tested.

I. Device Fabrication

An ITO coated glass piece (1 x 1cm^2 cut from large sheets purchased from Balzers. Switzerland) had a portion etched out with concentrated hydrochloric acid to remove the ITO and was cleaned and placed on a spin coater (CPS 10 BM, Semitec. Germany) and spun at 2000 rpm for 30 seconds, during which time the solution of the electroluminescent compound was dropped onto it dropwise by a pipette.

Alternatively the electroluminescent compound was vacuum evaporated onto the ITO coated glass piece by placing the substrate in a vacuum coater and evaporating the electroluminescent compound at I0^{-5} to I0^{-6} torr onto the substrate.

The organic coating on the portion which had been etched with, the concentrated hydrochloric acid was wiped with a cotton bud.

The coated electrodes were stored in a vacuum desiccator over calcium sulphate until they were loaded into a vacuum coater (Edwards, 10^{-6} torr) and aluminium top contacts made. The active area of the LED's was 0.08 cm2 by 0.1 cm^2 the devices were then kept in a vacuum desiccator until the electroluminescence studies were performed.

The ITO electrode was always connected to the positive terminal. The current vs. voltage studies were carried out on a computer controlled Keithly 2400 source meter.

Electroluminescence spectra were recorded by means of a computer controlled charge coupled device on Insta Spec photodiode array system model 77II2 (Oriel Co.. Surrey, England)

2. Photoluminescence Measurements

Photoluminescence was excited using 325mn line of Liconix 4207 NB, He/Cd laser. The laser power incident at the sample ($0.3mWcm^{-2}$) was measured by a Liconix 55PM laser power meter. The radiance calibration was carried out using Bentham radiance standard (Bentham SRS8, Lamp current 4,000A, calibrated by National Physical laboratories, England. The PL studies were carried out on samples or films. The Complexes of the examples were tested and the results shown in the Table and the Spectra attached as Figs. 2 to 6

Table

Example	PL %	λmax/nm	CIE		Colour
			x	y	
1	1.0	450	0.17	0.15	Purple
2	6.0	410,520	0.21	0.32	Greenish Blue
3	0.03	460	0.21	0.29	White
4	16	320,450			Purple
5	0.9	420	0.18	0.05	Purple

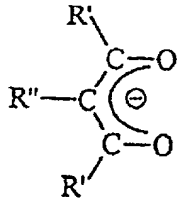
Claims

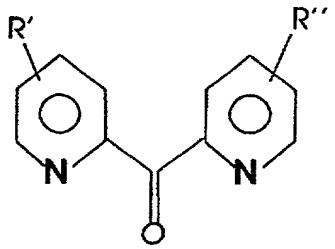
1. A photoluminescent compound which comprises an organic complex of a transition metal, a lanthanide or an actinide and an organic ligand which photoluminescent compound emits light in the blue or purplish blue spectrum.

2. An electroluminescent compound which comprises an organic complex of a lanthanide or an actinide and an organic ligand which electroluminescent compound emits light in the blue or purplish blue spectrum when an electric current is passed through it.

3. A compound as claimed in claim I or 2 which comprises a complex of thorium (IV), yttrium (III), gadolinium (III), europium (II), terbium(IV), cerium(IV) and cerium (III) or a mixture of one or more of these.

4. A compound as claimed in claim 1, 2 or 3 in which the ligand is selected from





or

where R' maybe the same or different at different parts of the molecule and each of R" and R' is a substituted or unsubstituted aromatic or heterocyclic ring structure or a hydrocarbyl or a fluorocarbon or R'' is fluorine or hydrogen or R'' is copolymerised with a monomer or is an alkyl group preferably a -C(CH3) group, or is selected from

TMHD, α,' α'', α''' tripyridyl, bathophen (4,7-diphenyl-I,I0-phenanthroline), crown ethers and cryptans.

5. A compound as claimed in claim 4 in which the ligand is selected from thorium (IV) bathophen, yttrium (III) tripyridyl and yttrium (III) TMHD, and europium (II) TMHD complexes.

6. $Eu(II)(TMHD)_2$.

7. A composition which comprises an inert polymer and from 5% to 95% by weight of an electroluminescent compound as claimed in any one of the preceding claims.

8. An electroluminescent device which comprises a transparent substrate on which is deposited an electroluminescent compound as claimed in any one of the preceding claims.

9. An electroluminescent device as claimed in claim 8 in which the transparent substrate comprises a conductive glass or plastic material which acts as the anode.

10. An electroluminescent device as claimed in claim 9 in which the transparent substrate comprises an indium tin oxide coated glass.

11. An electroluminescent device as claimed in any one of claims 8 to 10 in which there is a hole transporting layer deposited on the transparent substrate and the electroluminescent material is deposited on the hole transporting layer.

12. An electroluminescent device as claimed in claim 11 in which there is a hole transporting material mixed with the electroluminescent material in a ratio of 5 to 95% of the electroluminescent material to 95 to 5% of the hole transporting compound.

13. An electroluminescent device as claimed in claim 12 in which the hole transporting layer is an aromatic amine complex.

14. An electroluminescent device as claimed in claim 13 in which the hole transporting layer is poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD) or polyaniline.

15. An electroluminescent device as claimed in any one of claims 8 to 14 in which there is a metal anode in contact with the electroluminescent material.

16. An electroluminescent device as claimed in any one of claims 8 to 15 in which there is a layer of an electron injecting material between the cathode and the electroluminescent material layer

17. An electroluminescent device as claimed in any one of claims 8 to 16 in which an electron injecting material is mixed with the electroluminescent material and co-deposited with it.

18. An electroluminescent device as claimed in claim 16 or 17 in which the electron injecting material is a metal complex or oxadiazole or an oxadiazole derivative.

19. An electroluminescent device as claimed in claim 18 in which the electron injecting material is an aluminium quinolate or 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4 oxadiazole.

20. An electroluminescent device as claimed in any one of claims 8 to 19 in which there is a dye incorporated in the electroluminescent layer.

21. An electroluminescent device as claimed in 20 in which the dye is a fluorescent

laser dye or an electroluminescent laser dye.

22. An electroluminescent device as claimed in any one of the preceding claims 8 to 20 in which the anode is a metal.

23. An electroluminescent device as claimed in claim 22 in which the anode is a aluminium, magnesium, lithium, calcium or a magnesium silver alloy.

24. An electroluminescent device as claimed in any one of the preceding claims in which there are a plurality of layers of electroluminescent material.

25. An electroluminescent device as claimed in any one of the preceding claims in which the layer of electroluminescent material is formed of two or more different electroluminescent compounds.

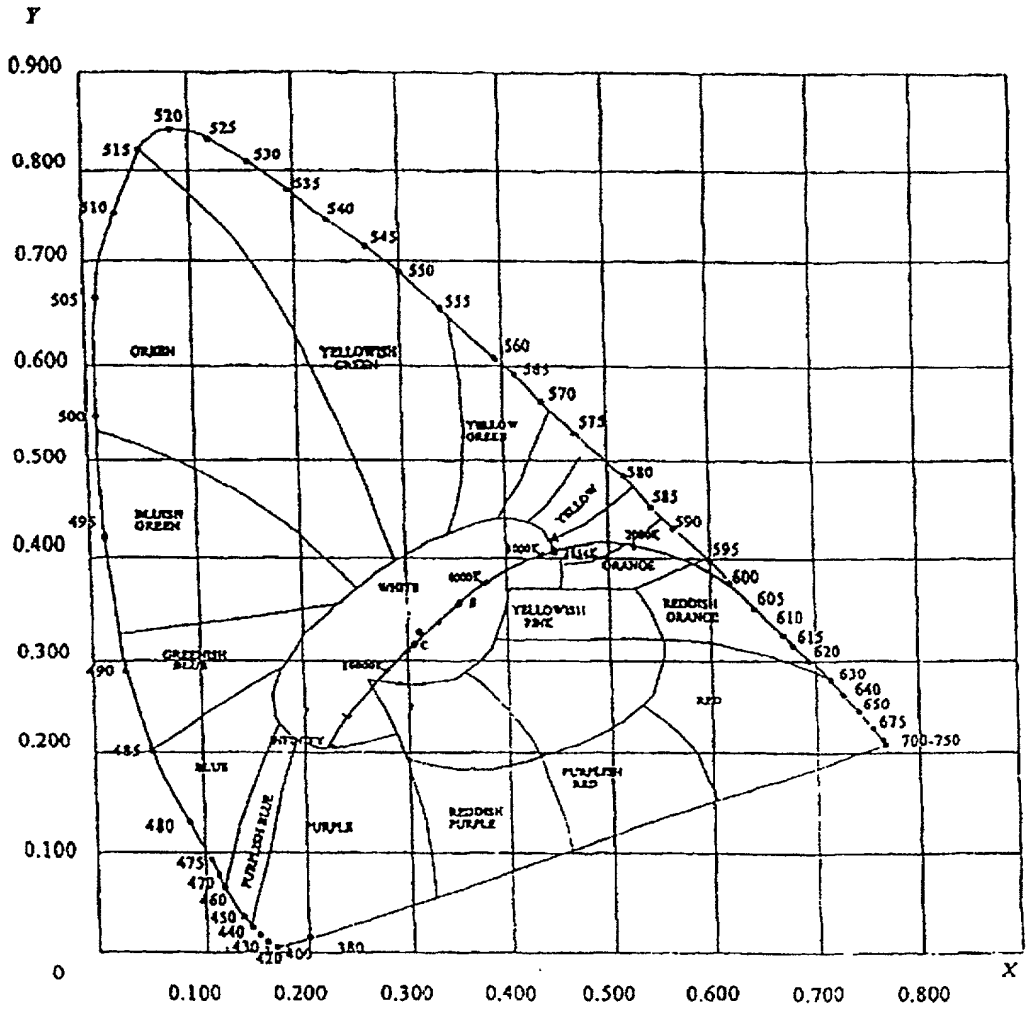


FIG. 1

CIE 1931 x,y chromacity diagram showing approximate position of perceived colours

Example 1

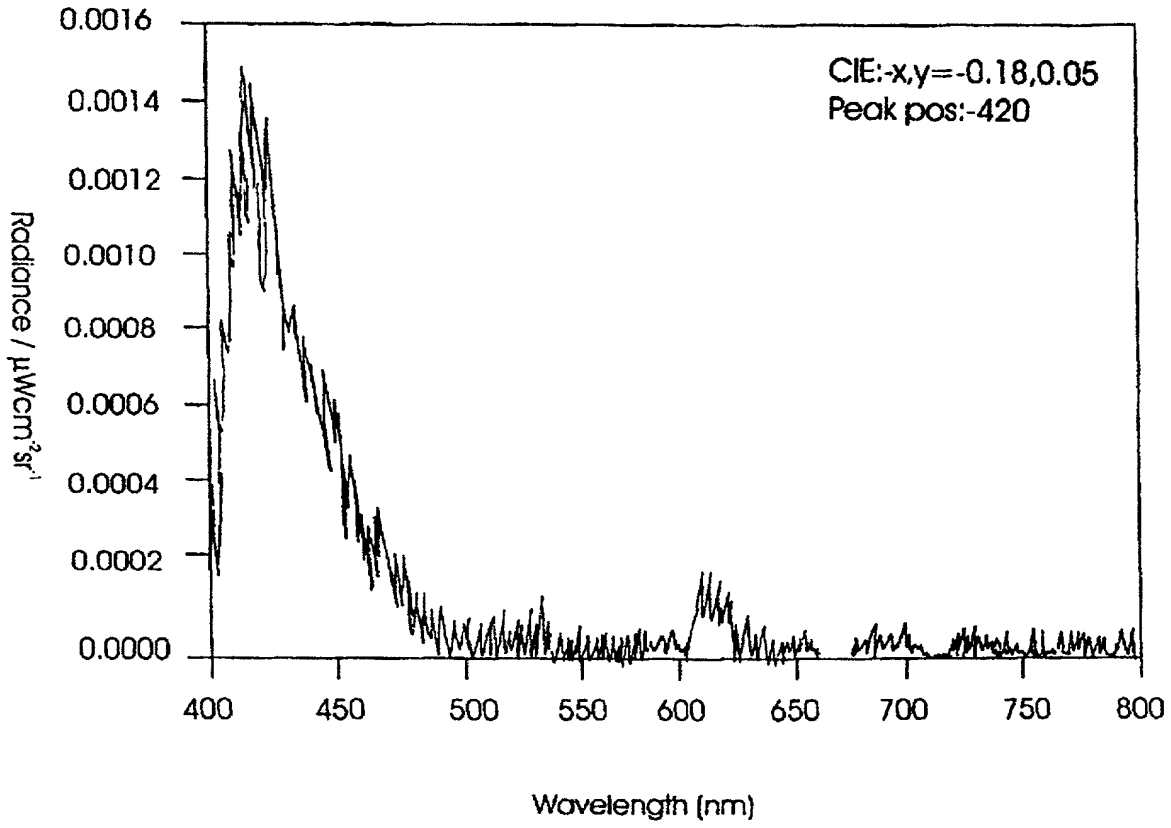


Fig. 2

Example 2

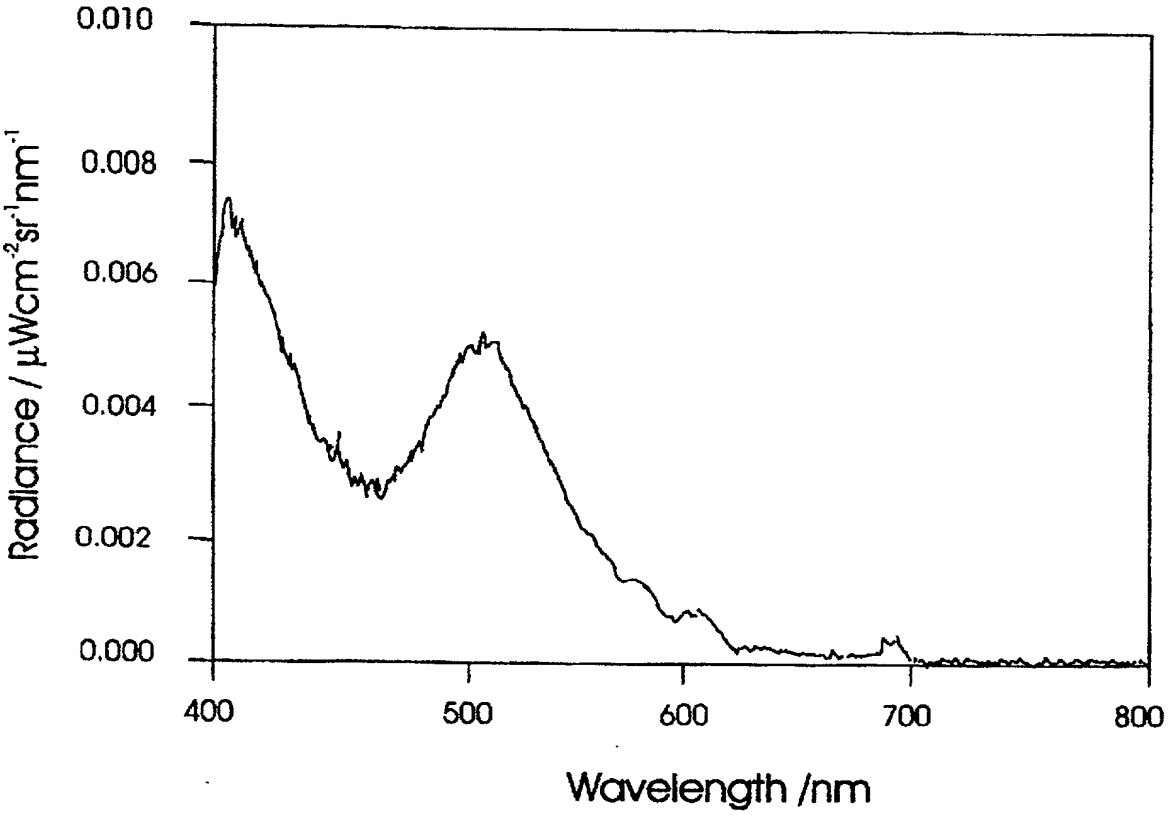


Fig. 3

Example 3

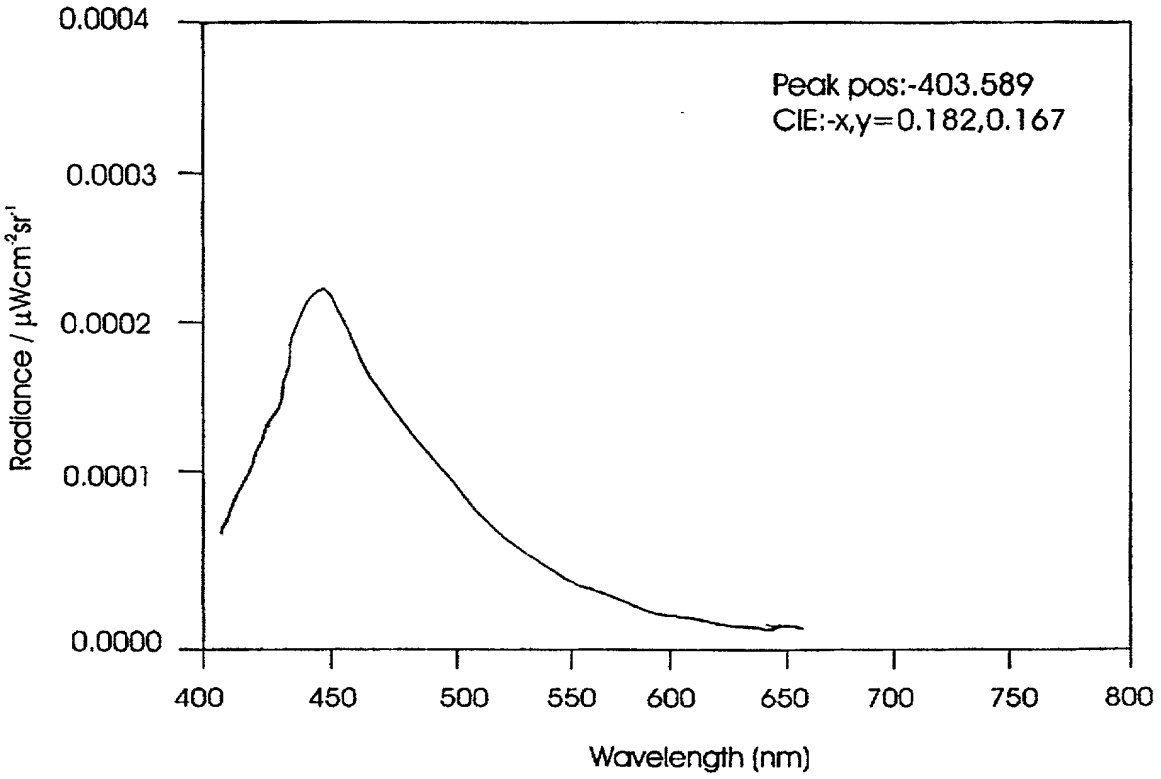


Fig. 4

Example 4

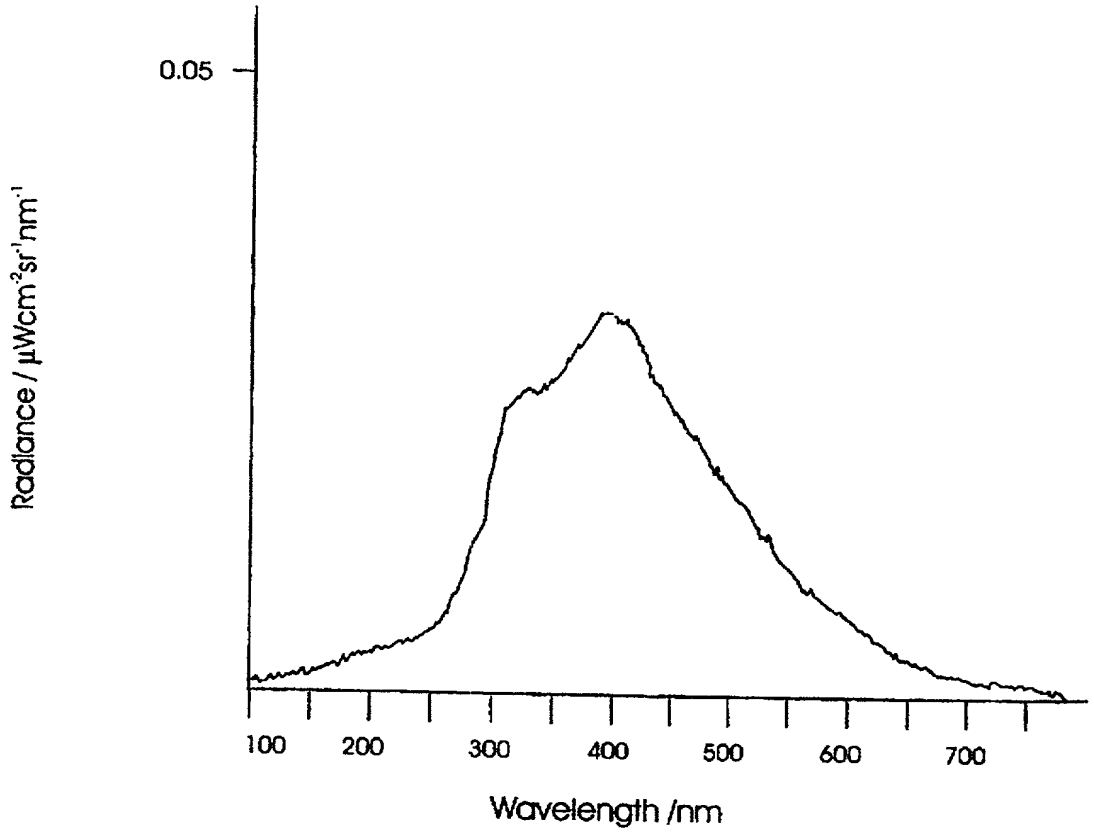


Fig. 5

Example 5

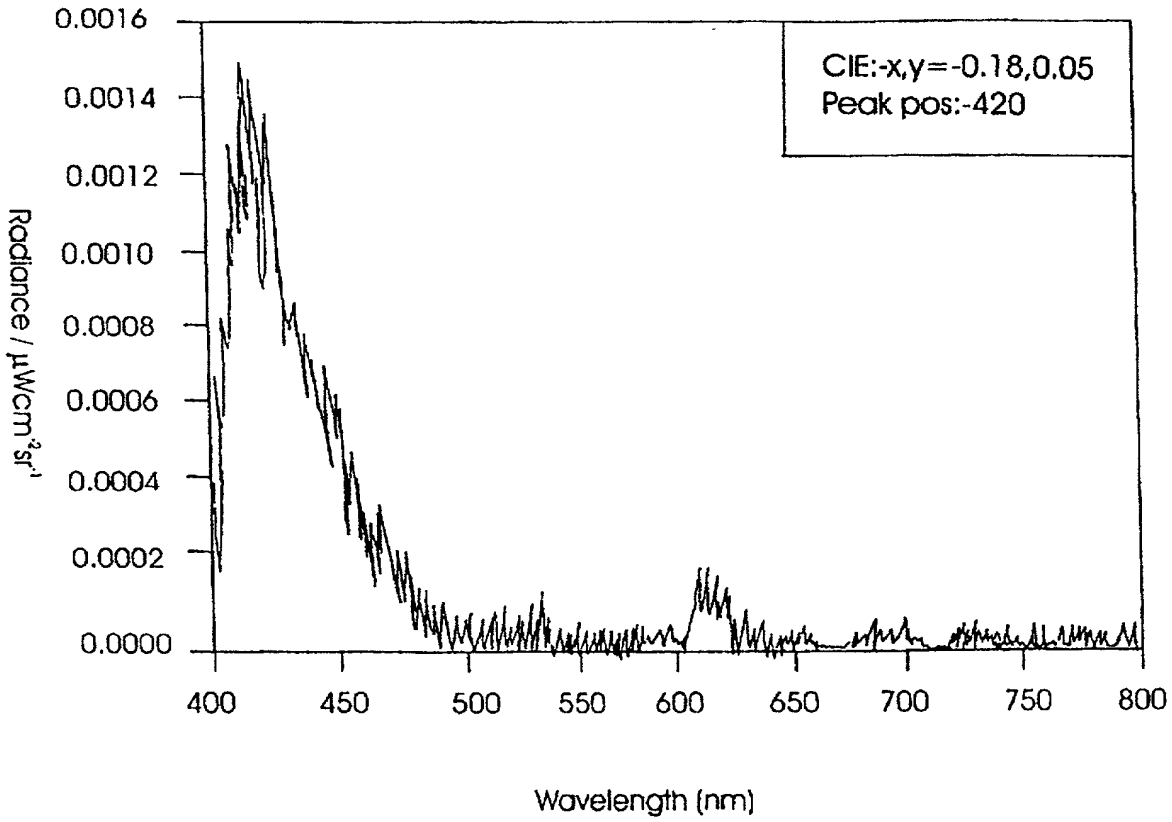


Fig. 6

Docket No.
UEL,008-US

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

ELECTROLUMINESCENT MATERIALS

the specification of which

(check one)

☐ is attached hereto.

☒ was filed on 2nd November 1999 as United States Application No. or PCT International Application Number PCT/GB99/03619

and was amended on _____
(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)			Priority Not Claimed
UK 98 23761.3	UK	2nd November 1998	☐
(Number)	(Country)	(Day/Month/Year Filed)	
			☐
(Number)	(Country)	(Day/Month/Year Filed)	
			☐
(Number)	(Country)	(Day/Month/Year Filed)	

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

(Application Serial No.)	(Filing Date)
(Application Serial No.)	(Filing Date)
(Application Serial No.)	(Filing Date)

I hereby claim the benefit under 35 U. S. C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112, I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C. F. R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. *(list name and registration number)*

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